

The convergence method to calculate particles fluxes in x rays spectrometry techniques. Application in nuclear compounds

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Abstract

A method to calculate particle fluxes applicable in most of the spectroscopy techniques is described. Flux intensities of backscattered or absorbed electrons and emitted photons are calculated using a method of convergence to solve the Invariant Embedding equations that are used to describe the particle trajectories inside a solid sample. Our results are found to be helpful to carry out a procedure for quantitative characterization using instrument such as Electron Probe Microanalysis or other probes. Examples of application to calculate the composition of ternary alloys are given and are compared with the same calculations using another procedure.

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I. INTRODUCTION

In most of the spectroscopy techniques, the signals produced by the different probes must be recorded and interpreted considering the different kinds of interactions between particles during its trajectory through an analyzed sample. This procedure involves hard calculations of fluxes of particles like electrons and/or photons, neutrons, protons etc. These fluxes must be obtained solving a transport problem where some particles produce other particles: as an example, electrons transport could produce electrons, photons or both of them as a consequence of their interactions with matter. The transport problem, in the spectroscopy area, is usually solved using Monte Carlo Calculation [1–4] or numerical solutions of the time-independent Boltzmann transport equation. Although considerably advances were made in the subject, most of the methods used in the practice have an empirical or semi empirical origin [5–10].

The quantitative determination of the unknown chemical composition of a material is an essential goal for its characterization. Among the spectroscopy techniques, the most used by researchers in materials science for compositional determinations is the Electron Probe Microanalysis (EPMA). EPMA is based on the detection of the characteristic X-rays emitted from a solid sample when a beam of electrons impinges on its surface with energies that typically vary between 2 and 50 keV. This instrument combines the capabilities of both the scanning electron microscope and an X-ray fluorescence spectrometer. The electrons of the beam are scattered as they penetrate within the sample producing backscattered electrons, secondary electrons, absorbed electrons and characteristic and continuum X-rays spectra. Quantitative microanalysis in EPMA is performed by comparing the characteristic X-ray intensity from one element in the sample with that from a reference standard containing a known amount of the same element, by means of the so called the K ratio,

$$K = \frac{I}{I_p} \quad (1)$$

Where I and I_p are the intensities of the characteristic X-rays emitted from the analyzed sample and that from a standard. [2, 11, 12]. The K ratio could be transformed into the mass concentration of the element of interest taking into account some correction factors due to processes undergone by the characteristic X-ray and by the electrons of the beam inside the sample. These processes involve scattering dynamics related to the sizes of the different atomic nuclei present in the sample and the standard, absorption, fluorescence.

The respective correction factors arising from each of these processes are called Z , A and F in the literature [2]. Then, the atomic fractions of the elements are obtained using the following expression:

$$K = \frac{I}{I_p} = ZAF \frac{C}{C_p} \quad (2)$$

Where C and C_p are the atomic fractions of the element of interest in the sample and in the standard.

The most common procedures to estimate the corrections are the so called *Matrix Correction* which are based on the calculation of the X-ray production as a function of the sample depth, $\varphi(\rho z)$ to estimate both the absorption and the atomic number corrections [2, 13, 14]. Most of the analytical expressions for the $\varphi(\rho z)$ functions used in commercial software packages are obtained from ad hoc models. Also the expressions for the $\varphi(\rho z)$ function and for the backscattering electron coefficients are considered as unrelated [15]. On the other side, in the literature there are theoretical models based on Boltzmann's transport equation to describe the phenomenon and to interpret the signals in EPMA [16, 17]. Although, this procedure has the disadvantage that there are solutions only for the one dimension problem, and little progress has been made on the 2-D or 3-D cases. Therefore this has a reduced practical application. Monte Carlo calculations are still the most worthwhile method of approaching theoretical problems in EPMA [5, 12, 18, 19]. However, depending on the accuracy or resolution required, running times can be computationally expensive.

In the last two decades a few authors demonstrated the efficiency of the Invariant Embedding (IE) method to describe the scattering of electrons inside a solid sample [20–27]. This method is useful to estimate the flux of particles through the interfaces in a material medium as a function of experimental parameters and the detected signals. It is, the IE method avoids calculations of fluxes inside the analyzed samples, thus gaining calculation advantages, and yields a set of equations for the detected fluxes as a function of the size of the analyzed sample. This set can be solved as an initial-value problem. Particularly, the backscattering problem was successfully solved by different authors using IE method [22, 28]. However, a procedure based on IE, useful for the experimentalist to perform chemical characterization, is still lacking.

In this article we focus on the calculation of the chemical compositions of a materials by using the experimental K ratios and theoretical X-ray intensities obtained with IE. Although the IE method is a powerful tool in order to simplify the transport equations, in

3-D transport problems the addition of degrees of freedom of the particles generates more complex mathematic. The Method of Convergence (MC) proposed here allows us to deduce closed approximated solutions for the 3-D IE equations for the detected X-rays intensities. These intensities can be calculated with no need of previous knowledge of the $\varphi(\rho z)$ function because the absorption corrections are already included in the equations. Here we propose two solutions for the IE equations with different levels of approximation. One called the *simple expression* and the other, more rigorous, called the *complete expression*. Comparison with other model proves that both solutions are a suitable tool for the calculation of intensities and concentrations.

II. THEORETICAL MODEL

The states ladder model and MC were are described in previous reports[25]. Basically, after impinging with energy E_0 , electrons can suffer two kinds of interactions: elastic or inelastic. Elastic collisions only cause deviations while inelastic ones only produce energy losses. Based on the assumption that it is allowed only one kind of inelastic collision with a constant energy loss of value ΔE , a discrete set of possible values is generated: E_1, E_2, E_3 and E_4 , for a five level system. For energies $E < E_4$, electrons are deactivated. Characteristic radiation can be generated exclusively by the inelastic collisions. Contributions $I_{01}, I_{02}, I_{03}, I_{04}$ to the total intensity are made by electrons backscattered with E_1, E_2, E_3 and E_4 , while contribution I_4 corresponds to the absorbed electrons. All these intensities can be calculated with the IE method. This method starts with the deduction of the integro-differential equation for each contribution to the intensity, whose derivation variable is the thickness τ of the sample. In the case of I_{α_0, β_1} , the intensity contribution for the electrons that impinge with E_0 and α_0 and are backscattered with E_1 and β_1 , the following equation is obtained:

$$\begin{aligned}
\frac{dI_{\hat{\alpha}_0, \hat{\beta}_1}(\tau)}{d\tau} = & \frac{g_0 R_{\hat{\alpha}_1, \hat{\beta}_1}(\tau) s_0}{\hat{\alpha}_0} \\
& + \frac{g_0 R_{\hat{\alpha}_0, \hat{\beta}_0}(\tau) s_0}{\hat{\beta}_1} - I_{\hat{\alpha}_0, \hat{\beta}_1} \left(\frac{2(s_0 + \sigma_0)}{\hat{\alpha}_0} + \mu \right) + \\
& \int_{\varphi} \frac{d\sigma_{\hat{\alpha}_0, \hat{\varphi}} I_{\hat{\varphi}_0, \hat{\beta}_1}(\tau)}{\hat{\alpha}_0} + \int_{\epsilon} \frac{d\sigma_{\hat{\epsilon}_1, \hat{\beta}_0} I_{\hat{\alpha}_0, \hat{\epsilon}_1}(\tau)}{\hat{\epsilon}_1} + \\
& \int_{\epsilon} \frac{d\sigma_{\hat{\epsilon}_1, \hat{\beta}_0} I_{\hat{\alpha}_0, \hat{\epsilon}_1}(\tau)}{\hat{\epsilon}_1} + \int_{\epsilon} \int_{\eta} \frac{d\sigma_{\hat{\epsilon}_0, \hat{\eta}_0} R_{\hat{\alpha}_0, \hat{\epsilon}_0}(\tau) I_{\hat{\eta}_0, \hat{\beta}_1}(\tau)}{\hat{\epsilon}_0} + \\
& \int_{\gamma} \int_{\delta} \frac{d\sigma_{1, \gamma, \delta} R_{\hat{\delta}_1, \hat{\beta}_1}(\tau) I_{\hat{\alpha}_0, \hat{\gamma}_1}(\tau)}{\hat{\gamma}_1}.
\end{aligned} \tag{3}$$

Were the simplified notation $\hat{\alpha}_j = \cos(\alpha_j)$ and $\hat{\beta}_j = \cos(\beta_j)$ is used for the incident and emerging director cosines of the polar angles of the electrons, measured with respect to the normal of the the sample surface. The index $j = 0, 1, ..$ labels the energy states values, σ_j and s_j are the elastic and inelastic cross sections, respectively; φ , ϵ and δ are the scattering angles due to single interactions inside the sample, and g_j is the electrons efficiency of X-ray production per collision.

Using the MC it is possible to obtain two approximate solutions for the last equation: the complete form, i.e., the formal solution, and the simple form, used initially as an auxiliary expression to solve the equations of the lower levels. These approximate solutions tend to an exact solution for $\tau \rightarrow \infty$. The simple form is:

$$I_{\hat{\alpha}_0, \hat{\beta}_1}(\infty) = \frac{g_0 \hat{\alpha}_0 \hat{\beta}_1 s_0}{(\lambda + 1)(s_1 \hat{\alpha}_0 + s_0 \hat{\beta}_1) + \mu \hat{\alpha}_0 \hat{\beta}_1} \frac{(1 + \lambda) \Gamma_1}{\lambda J_0} \tag{4}$$

Where $J_0 = \lambda + 2 - \alpha \Gamma_1$; $\Gamma_1 = \lambda = 2 - 2\sqrt{\lambda + 1}$ and $\lambda = \sigma_0/s_0$.

In order to study the applicability range of the solutions, chemical composition calculations made with the simple and the complete expressions are presented. The factor of generation gj it calculated starting from Bethe's expression for L_k lines, and an energy level E_j , for the ionized element in the compound:

$$g_{kj} = 12.67210^{-8} \frac{\rho_k N_A}{A_k E_j I_k} L_n \left(\frac{E_j}{1.610^{-16} l_k} \right) \sqrt{\frac{2E_j}{m}} \tag{5}$$

Where ρ_k is the partial density, A_k is the atomic number and m is the mass of the electron. Parameters such as the partial densities, ionization energies or attenuation coefficients are obtained from tables. The cross sections are calculated from classic expressions as functions

of the partial densities. In this model, it is considered that the beam electrons interact with atoms in the compound, whose cross sections is the pondered average of the cross sections of each element. The procedure is as follows: 1) The summation of the measured K ratios is normalized. 2) This normalized values are considered as the initial atomic concentrations. 3) With these new initial atomic fractions, the K ratios are recalculated with the simple and the complete MC expressions, and the results are compared with the measured values. 4) In successive calculations, the atomic fractions of both expressions are adjusted in order to fit the theoretical and experimental K ratios. The final values for the concentrations are those that yield the best fit. The calculation was performed for the line L_α of Zr and K_α of Fe.

III. CHARACTERISTIC INTENSITIES. SIMPLE EXPRESSIONS.

We proceed to calculate I01, I02, I03, I04 and I4 in order to obtain $I = \sum_i I0i$

The simple expressions for the Intensities are:

$$I01(\infty) = \int_0^1 \frac{4g_0a_0s_0\hat{\beta}_1(1+\lambda)T_1d\hat{\beta}_1}{\{(\lambda+1)(s_1\hat{\alpha}_0 + s_0\hat{\beta}_1) + \mu\hat{\alpha}_0\hat{\beta}_1\}\lambda J_0} \quad (6)$$

$$I02(\infty) = \int_0^1 \frac{8g_2a_0s_0(1+\lambda)T_1W_0\hat{\beta}_2d\hat{\beta}_2}{[(s_2\hat{\alpha}_0 + s_0\hat{\beta}_2)(1+\lambda) + \mu\hat{\alpha}_0\hat{\beta}_2]\lambda J_0^3} \quad (7)$$

$$I03(\infty) = \int_0^1 \frac{16g_3a_0s_0(1+\lambda)T_1\hat{\beta}_3d\hat{\beta}_3\{W_0R_0 + b_0c_0Z_0(\lambda+2)\}}{(s_3\hat{\alpha}_0 + s_0\hat{\beta}_3)(1+\lambda) + \mu\hat{\alpha}_0\hat{\beta}_3\lambda J_0^4} \quad (8)$$

$$\begin{aligned} I04(\infty) = & \int_0^1 \frac{32g_4a_0s_0T_1(\lambda+1)\hat{\beta}_4}{\{\lambda J_0^6[s_4\hat{\alpha}_0 + s_0\hat{\beta}_4]\}} \\ & \times \{b_0c_0J_0Z_0(d_0V_0 + \lambda + 2) + W_0(a_0b_0T_1 + J_0) + \\ & a_0b_0T_1\{W_0R_0 + c_0Z_0[b_0(\lambda+2) + d_0V_0 + W_0]\}\}d\hat{\beta}_4 \end{aligned} \quad (9)$$

where

$$Z_0 = \frac{a_0(b_0 - 2)T_1 + 2\lambda + 4}{\lambda + 2 + 2\sqrt{\lambda + 1} - a_0T_1} \quad (10)$$

$$W_0 = (2 + \lambda)(b_0 + 1) - a_0T_1 \quad (11)$$

IV. ASSESSMENT OF THE RESULTS: ZR-BASED ALLOYS

In order to compare the calculated K ratios and concentrations between MC and PAP [29–31] results, we used experimental data obtained with an analytical microprobe of electrons CAMECA SX50 with 20 kV of accelerating potential from samples of ternary metallic alloys of the systems Zr-Sn-Fe and Zr-Sn-Nb. For the calibration, high purity standards of each of the analyzed elements were used. The proposed method was applied to a set of fourteen measurements on these alloys. The nominal atomic composition of the Zr-Sn-Fe sample is: Zr 62.5 - Sn 25.0 - Fe 12.5. It was thermally treated to 800C for 2000 hours. Three phases are detected in this sample: two binary intermetallic compounds (ZrFe_2 and Zr_5Sn_3) and a ternary compound $\text{Zr}_6\text{Sn}_2\text{Fe}$. At first, the atomic composition was estimated using the PAP procedure. On the other hand, the theoretical results of section II and III were used to estimate the theoretical K ratios and the atomic composition with the complete and the simple expressions for the characteristic intensities. The results are shown in tables I and II. They show comparisons between PAP concentrations and results obtained with the simple and the complete 3D-IE expressions, for the Zr_5Sn_3 and $\text{Zr}_6\text{Sn}_2\text{Fe}$ phases. For each: element of interest, measured K ratio (k), concentrations from the PAP method (at(PAP)), concentrations from 3D-IE simple equation (at(3Ds)), concentrations from 3D-IE complete equation (at(3Dc)), K ratios from 3D-IE simple equation (ks), K ratios from 3D-IE complete equation (kc). Each table contains different sets of values, corresponding to different measuring points in the same phase. The first rows of each set of values indicate the PAP, 3Ds and 3Dc estimations of the oxygen concentration, as this elements is always present at the surface of these alloys.

On the other hand, the nominal atomic compositions of the Zr-Sn-Nb samples are: Zr 70.0 - Sn 5.0 - Nb 25.0 for the first one, and Zr 65.0 - Sn 10.0 - Nb 25.0 for the other one. They were thermally treated to 950C for 2900 hours. Table III shows comparisons of the theoretical results in this article and experimental data, in the same way as Tables I and II.

TABLE I: Theoretical calculations (IE) for the K ratios of the elements (E) in the Zr_5Sn_3 phase of the Zr-Sn-Fe alloy. The 3D-IE results obtained with the complete (c) and the simple (s) expression are compared with PAP calculation.

		3.16% O	2.6% O	15.1% O		
	K	%at(PAP)	%at(3Ds)	% at (3Dc)	$K_s(t)$	$K_c(t)$
Zr	0.4498	61.3297	53.1501	60.9755	0.4460	0.4791
Sn	0.3303	38.2826	46.4461	38.4317	0.3268	0.3418
Fe	0.0020	0.378	0.4038	0.5928	0.0020	0.0020
		6.28% O	1.1% O	13.1% O		
Zr	0.4527	61.7237	53.93	61.4206	0.4573	0.4883
Sn	0.3336	37.9375	45.6761	38.0250	0.3243	0.3486
Fe	0.0018	0.3387	0.3557	0.5544	0.0018	0.0019

V. DISCUSSION

Calculations performed with the simple and the complete expressions obtained with MC are compared with results of the PAP method. The fitting is acceptable keeping in mind the conditions of calculation and the fact that the intensity expressions were deduced from approximate expressions for the transport coefficients of the electrons. Thereby, the intensities are a second stage of approximation. In the Zr-Sn-Fe results, the presence of oxygen changes the composition of the phases in a significant way. Oxygen diminishes the K ratios of the other elements, in comparison with non-oxidized samples. In relation to the phase Zr_5Sn_3 , it is necessary to remark that the results yielded by the complete MC expressions fit the PAP concentrations better than the simple ones. However, the complete expressions overestimate the oxygen concentrations at the surface. In relation to the MC calculated K ratios, the discrepancy is less than 5%, with respect to the PAP values. In general, for this compound, the simple MC expressions underestimate the Zr and overestimate the Sn, in comparison with PAP and complete MC expressions. It is necessary to emphasize that the equations are valid if the ratio $\psi = \mu/s_0$ can be considered as negligible with respect to unity. In the calculations, Ψ values are: 0.05% for Fe, 0.18% for Zr and 0.14% for Sn. It means that the MC expressions are at the limit of its applicability for the present selection

TABLE II: Theoretical calculations (3D-IE) for the K ratios of the elements (E) and its atomic fractions in the $\text{Zr}_6\text{Sn}_2\text{Fe}$ phase of the Zr-Sn-Fe alloy. The 3D-IE results are compared with PAP calculation.

		1.14% O	7.0% O	7.0% O		
	K	%at(PAP)	%at(3Ds)	% at (3Dc)	$K_s(t)$	$K_c(t)$
Zr	0.5716	67.36	61.2063	65.294	0.5561	0.5982
Sn	0.2120	22.7	27.7656	21.6365	0.2090	0.2205
Fe	0.0604	9.925	11.028	13.071	0.0605	0.0625
		1.47% O	5.5% O	7.0% O		
Zr	0.5763	67.43	62.1693	65.3326	0.5692	0.5990
Sn	0.2115	22.5	27.2487	21.5054	0.2065	0.2193
Fe	0.0616	10.05	10.5820	13.1720	0.0585	0.0631
		1.18%O	7.4%O	15.4%		
Zr	0.5102	65.0721	58.1445	63.4064	0.4985	0.5228
Sn	0.2680	30.1344	36.6928	29.8788	0.2621	0.2797
Fe	0.0272	34.8033	5.16274	6.7148	0.0268	0.0285
		2.13%O	4.0%O	7.5%		
Zr	0.5625	66.5781	60.6771	64.5946	0.5561	0.5870
Sn	0.2230	23.7969	29.1667	22.7027	0.2217	0.2304
Fe	0.0585	9.6352	10.1562	12.7027	0.0562	0.0603
		0.21%O	2.3% O	9.0%		
Zr	0.5629	66.4305	60.0338	64.4540	0.5783	0.5784
Sn	0.2292	24.3489	29.9064	23.3171	0.2340	0.2341
Fe	0.0562	9.2206	10.0596	12.2288	0.05729	0.0573

TABLE III: Theoretical calculations for K_α ratios of elements (E) in the ZrSnNb alloy. .

	K	%at(PAP)	%at(3Ds)	% at (3Dc)	$K_s(t)$	$K_c(t)$
Zr	0.6071	64.50	58.92	59.02	0.6129	0.5946
Sn	0.0220	2.59	1.74	2.44	0.0217	0.0228
Nb	0.3253	32.91	39.34	38.54	0.3275	0.3155
Zr	0.616	66.03	63.62	61.32	0.6337	0.6034
Sn	0.0678	7.81	5.72	6.74	0.0696	0.0671
Nb	0.2552	26.17	31.66	31.94	0.2556	0.2529
Zr	0.5957	64.88	59.02	59.02	0.6126	0.5934
Sn	0.0271	3.26	2.16	2.84	0.0269	0.0269
Nb	0.3076	31.86	38.82	38.14	0.3224	0.3115
Sample 2						
Zr	0.7291	78.39	74.77	74.77	0.7464	0.7309
Sn	0.0380	4.46	3.21	4.09	0.0325	0.0382
Nb	0.1677	17.14	22.02	21.14	0.1746	0.1629
Zr	0.6931	70.82	67.01	67.01	0.6831	0.6661
Sn	0.0294	3.51	2.99	2.99	0.0292	0.0278
Nb	0.2489	25.67	30.00	30.0	0.2487	0.2390
Zr	0.7071	73.64	68.78	68.10	0.6993	0.6762
Sn	0.0269	3.10	2.20	2.90	0.0268	0.0269
Nb	0.2344	23.26	29.02	29.0	0.2351	0.2304
Zr	0.6849	72.92	68.10	68.10	0.6932	0.6762
Sn	0.0277	3.25	2.25	2.90	0.0274	0.0269
Nb	0.2352	23.255	29.65	29.0	0.2405	0.2304

of parameters. In relation to the oxygen, the simple MC expressions overestimate its concentration in comparison with PAP. The results for $\text{Zr}_6\text{Sn}_2\text{Fe}$ show the same behavior that those of the previous phase in the sense that there is a better fit between concentrations calculated with the complete MC expressions and PAP in comparison with the simple ones. These expressions yield results for the K ratios that differ from the experimental values in an amount that ranges from 2 to 5 %. In general, the theoretical K ratios fit to experimental values within less than 3 %, with an overestimation in the results of the simple MC expressions. The ψ ratios for this phase are: 0.05 for Fe, 0.17 for Zr and 0.14 for Sn, so the MC expressions are at its limit of application. With regard to oxygen, PAP concentrations are about 1% while MC values range from 7 to 15%. In this phase the discrepancies in the concentrations of Zr, Sn and Fe between PAP and MC range from 2 to 5%. In the case of Zr-Sn-Nb, the fit between K ratios is better, but the discrepancy between PAP and MC concentrations is higher. Both MC expressions underestimate the Zr and overestimate the Nb in comparison with PAP results. There is no oxygen at the surface of this compound, and the Ψ ratios are: 0.12% for Nb, 0.16% for Zr and 0.17% for Sn. It is important to remark that the inelastic-elastic correlation determines the ionization rate and the $\phi(\rho z)$ function. Thereby, the intensities depend on such correlation, which is related to the λ factor, this is to say, to the ratio between the inelastic and elastic cross sections. Our calculations were performed basically to verify the internal coherence of the 3D model and the MC expressions. This means that parameters such as cross-sections, attenuation coefficients, ionization energies, etc are fixed values taken from tables, and the only adjustable parameter is the Emin value, i.e., the minimum energy lost per inelastic collision, whose magnitude was selected to fit the backscattering and absorption coefficients of the electrons. Bearing in mind this fact, the presented results for concentrations and K ratios are quite satisfactory and indicate that the 3D model and the MC expressions are appropriate for the theoretical representation of the interaction phenomena in EPMA, and thereby, for the characterization of compounds by means of this or similar techniques.

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Appendix A: Determination of the function $I_{\alpha_0, \beta_1}(\infty)$. Method of Convergence.

Considering in the integral-differential equation for $I_{\alpha_0, \beta_1}(\infty)$, Eq.(3) the six first terms equals to a constant I1:

$$\begin{aligned}
 I1 = & \frac{g_0 R00_{\alpha\beta} s_0}{\hat{\beta}_1} \\
 & + \frac{g_0 R11_{\alpha\beta} s_0}{\hat{\alpha}_0} + \int_{\varphi} \frac{d\sigma_{0\alpha\beta} I01_{\varphi\beta}}{\hat{\alpha}_0} \\
 & + \int_{\xi} \frac{d\sigma_{1\xi\beta} I01_{\alpha\xi}}{\hat{\xi}_1} + \int_{\gamma} \int_{\delta} \frac{d\sigma_{1\gamma\delta} I01_{\alpha\gamma} R11_{\delta\beta}}{\hat{\gamma}_1} + \\
 & \int_{\zeta} \int_{\eta} \frac{d\sigma_{0\xi\eta} R00_{\alpha\zeta} I01_{\eta\beta}}{\hat{\zeta}_0}
 \end{aligned} \tag{A1}$$

Eq.(3) is now written as

$$\frac{dI_{\alpha_0, \beta_1}(\tau)}{d\tau} = I1 - I_{\alpha_0, \beta_1}(\tau) \left(\frac{s_0 + \sigma_0}{\hat{\alpha}_0} + \frac{s_1 + \sigma_1}{\hat{\beta}_1} + \mu \right) \tag{A2}$$

The solution for $\tau \rightarrow \infty$ is:

$$I_{\alpha_0, \beta_1}(\infty) = \frac{I1 \hat{\alpha}_0 \hat{\beta}_1}{\hat{\alpha}_0(s_1 + \sigma_1) + \hat{\beta}_1(s_0 + \sigma_0) + \mu \hat{\alpha}_0 \hat{\beta}_1} \tag{A3}$$

Let us rename the term $I1 \times \hat{\alpha}_0 = B1$, were $B1 = B1(\hat{\alpha}_0, \hat{\beta}_1)$:

$$I_{\alpha_0, \beta_1}(\infty) = \frac{B1 \hat{\beta}_1}{\hat{\alpha}_0(s_1 + \sigma_1) + \hat{\beta}_1(s_0 + \sigma_0) + \mu \hat{\alpha}_0 \hat{\beta}_1} \tag{A4}$$

Equation [A1] contains four integrals:

$$\int_{\varphi} \frac{d\sigma_{0\alpha\beta} I_{\varphi_0, \beta_1}(\infty)}{\hat{\alpha}_0} \tag{A5}$$

$$\int_{\xi} \frac{d\sigma_{1\xi\beta} I_{\alpha_0, \xi_1}(\infty)}{\hat{\xi}_1} \tag{A6}$$

$$\int_{\gamma} \int_{\delta} \frac{d\sigma_{1\gamma\delta} I_{\alpha_0, \gamma_1}(\infty) R_{\delta_1 \beta_1}}{\hat{\gamma}_1} \tag{A7}$$

$$\int_{\zeta} \int_{\eta} \frac{d\sigma_{0\xi\eta} R_{\alpha_0 \zeta_0} I_{\eta_0 \beta_0}}{\hat{\zeta}_0} \tag{A8}$$

These integrals are solved by replacing $I_{\alpha_0, \beta_1}(\infty)$ by the expression [A4] obtained with the MC. The functions of Elastic Backscattering R00 are replaced by its normalized simple expression, deduced with the MC[reference]. Replacing at [A1], solving, multiplying for α_0 and replacing $\rightarrow \Psi_0 = \mu/s_0$ and $\Psi_1 = \mu/s_1$ we obtain.

$$\begin{aligned}
B1 &= \frac{2g_0a_0s_0T_1}{\lambda} + \frac{\lambda s_0B1\hat{\beta}_1}{2s_1[\lambda+1+\Psi_1\hat{\beta}_1]} \\
&\times \ln\left(\frac{s_1}{\hat{\beta}_1s_0} + 1 + \frac{\Psi_0}{\lambda+1}\right) + \frac{a_0s_1B1T_1\hat{\alpha}_0\hat{\beta}_1}{s_0[\lambda+1+\Psi_0\hat{\alpha}_0]} \\
&\times \ln\left(\frac{s_0}{\hat{\alpha}_0s_1} + 1 + \frac{\Psi_1}{\lambda+1}\right) \ln\left(1 + \frac{1}{\hat{\beta}_1}\right) + \frac{a_0s_0B1T_1\hat{\alpha}_0\hat{\beta}_1}{s_1[\lambda+1+\Psi_1\hat{\beta}_1]} \\
&\times \ln\left(\frac{s_1}{\hat{\beta}_1s_0} + 1 + \frac{\Psi_0}{\lambda+1}\right) \ln\left(1 + \frac{1}{\hat{\alpha}_0}\right) \\
&+ \frac{\lambda s_1B1\hat{\alpha}_0}{2s_0[\lambda+1+\Psi_0\hat{\alpha}_0]} \ln\left(\frac{s_0}{\hat{\alpha}_0s_1} + 1 + \frac{\Psi_1}{\lambda+1}\right)
\end{aligned}$$

Substituting this results in [A4] we obtain the following expression for I01:

$$\begin{aligned}
I01_{\alpha\beta}(\infty)^c &= \frac{\hat{\beta}_1}{\hat{\alpha}_0(s_1 + \sigma_1) + \hat{\beta}_1(s_0 + \sigma_0) + \mu\hat{\alpha}_0\hat{\beta}_1} \\
&\left\{ \frac{2g_0a_0s_0T_1}{\lambda} + \lambda s_0\kappa_0g_0 \left\{ \frac{s_0\lambda\hat{\beta}_1}{2s_1[\lambda+1+\Psi_1\hat{\beta}_1]} \ln\left(\frac{s_1}{\hat{\beta}_1s_0} \right. \right. \right. \\
&+ 1 + \frac{\Psi_0}{\lambda+1} \left. \left. \left. + \frac{a_0s_1T_1\hat{\alpha}_0\hat{\beta}_1}{s_0[\lambda+1+\Psi_0\hat{\alpha}_0]} \ln\left(\frac{s_0}{\hat{\alpha}_0s_1} + 1 + \frac{\Psi_1}{\lambda+1}\right) \right. \right. \right. \\
&\times \ln\left(1 + \frac{1}{\hat{\beta}_1}\right) + \frac{a_0s_0T_1\hat{\alpha}_0\hat{\beta}_1}{s_1[\lambda+1+\Psi_1\hat{\beta}_1]} \ln\left(\frac{s_1}{\hat{\beta}_1s_0} + 1 + \frac{\Psi_0}{\lambda+1}\right) \\
&\times \ln\left(1 + \frac{1}{\hat{\alpha}_0}\right) + \frac{\lambda s_1\hat{\alpha}_0}{2s_0[\lambda+1+\Psi_0\hat{\alpha}_0]} \ln\left(\frac{s_0}{\hat{\alpha}_0s_1} + 1 + \frac{\Psi_1}{\lambda+1}\right) \left. \right\}
\end{aligned} \tag{A9}$$

Where the super index "c" it refer to "complete".

Let us now substitute $B1 = \kappa_0g_0\lambda s_0$ at [A4] and a simple expression of I01 is written as:

$$I01_{\alpha\beta}(\infty)^s = \frac{\kappa_0g_0\lambda s_0\hat{\beta}_1}{\hat{\alpha}_0(s_1 + \sigma_1) + \hat{\beta}_1(s_0 + \sigma_0) + \mu\hat{\alpha}_0\hat{\beta}_1}$$

Where the super index s it refer to *simple*. In order to determine κ_0 we use the equality:

$$\int_0^1 \int_0^1 I01_{\alpha\beta}(\infty)^s d\hat{\alpha}_0 d\hat{\beta}_1 = \int_0^1 \int_0^1 I01_{\alpha\beta}(\infty)^c d\hat{\alpha}_0 d\hat{\beta}_1$$

Substituting [A9] and [A10] in [A11], considering $s_0 = s_1$ and $\mu \ll s_i$,

$$\begin{aligned}
\int_0^1 \int_0^1 \frac{\kappa_0 g_0 \lambda d\hat{\alpha}_0 d\hat{\beta}_1}{(\lambda+1)(\hat{\alpha}_0+\beta_1)} &= \int_0^1 \int_0^1 \frac{g_0 \hat{\beta}_1}{(\lambda+1)(\hat{\alpha}_0+\beta_1)} \\
&\left\{ \frac{2a_0 T_1}{\lambda} + \kappa_0 \lambda \left\{ \frac{a_0 T_1 \hat{\alpha}_0 \hat{\beta}_1}{\lambda+1} \ln\left(1 + \frac{1}{\hat{\beta}_1}\right) \ln\left(\frac{1}{\hat{\alpha}_0} + 1\right) \right. \right. \\
&\quad \left. \left. + \frac{a_0 T_1 \hat{\alpha}_0 \hat{\beta}_1}{\lambda+1} \ln\left(\frac{1}{\hat{\beta}_1} + 1\right) \ln\left(1 + \frac{1}{\hat{\alpha}_0}\right) \right. \right. \\
&\quad \left. \left. \frac{\lambda \hat{\alpha}_0}{2(\lambda+1)} \ln\left(\frac{1}{\hat{\alpha}_0} + 1\right) + \right. \right. \\
&\quad \left. \left. \frac{\lambda \hat{\beta}_1}{2(\lambda+1)} \ln\left(\frac{1}{\hat{\beta}_1} + 1\right) \right\} \right\} d\hat{\alpha}_0 d\hat{\beta}_1
\end{aligned} \tag{A10}$$

Solving the integrals κ_0 is written as:

$$\kappa_0 = \frac{4a_0(\lambda+1)T_1}{\lambda^2 J_0} \tag{A11}$$

Substituting [A13] in [A9] and [A10], the simple and complete expressions for I01 are obtained respectively.

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